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Exciton dynamics in a novel high-yield GaInP quantum-wire array

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We report the carrier dynamics in a spontaneously organized array of quantum wires grown by a novel technique that involves strain induced lateral ordering (SILO). Our cw-photoluminescence (PL) measurements reveal a very strong optical anisotropy associated with these wires, while the time-resolved PL measurements demonstrate a very interesting carrier dynamics due to localization of excitons and slow interwire scattering. The high quality and freedom from defects of the SILO multiple quantum wire array are nicely borne out by the long decay photoluminescence times (~ 4 ns). © 1996 American Institute of Physics. [S0003-6951(96)00341-5]

The unique physical properties of quantum wires (QWR) are expected to give rise to improved performance characteristics for a multitude of optoelectronic devices. For example, quantum wire devices are expected to benefit from an enhanced light-matter interaction, a sharply peaked density of states and orders of magnitude suppression of impurity,¹ and optical phonon scattering.² After more than a decade of effort, techniques for growing quantum wires are still in a stage of infancy as compared to those for quantum wells. While there have been successful efforts to grow structures that demonstrate theoretically predicted one-dimensional effects, the stringent requirements demanded of wire structures for device purposes have not yet been achieved.³ Quantum wire structures grown by conventional methods of patterning, etching, or composition modulation typically suffer from one or more of the following shortcomings: insufficient confinement, large wire-width, increased surface defects, and/or low density of the active region. Recently, however, a novel technique of wire growth has been demonstrated⁴ that appears to be better suited for improving many of these problems. This growth process, involving strain induced lateral ordering (SILO), results in the formation of quantum wires with a periodicity of about 150–200 Å. The SILO growth technique involves growing a short period superlattice of incommensurate bilayers of the constituent compound semiconductors ($\text{GaP}_{2-\delta}/\text{InP}_{2+\delta}$ or $\text{GaAs}_{2-\delta}/\text{InAs}_{2+\delta}$) on a [001]-oriented GaAs substrate. The resulting elastic stress and strain fields have been shown⁵ to render an alloy with lateral composition modulation more thermodynamically stable, once it has been initiated, than a random homogeneous alloy. The result is a spontaneously arranged array of In-rich GaInP wires with Ga-rich GaInP barriers providing carrier confinement in the growth plane. Confinement in the growth direction is provided by the lattice-matched GaInP layers. In order to characterize the physical properties, and potential technological usefulness, of these quantum wire

structures, we have studied the photoluminescence (PL) and carrier dynamics in SILO grown GaInP multiple quantum wire arrays.

Shown in Fig. 1(a) is an idealized depiction of the sample structure. The SILO wires are grown sandwiched between GaInP buffer layers lattice matched to the GaAs substrate. Details of sample growth are published elsewhere.⁶ The approximate band gaps and the dimensions of all the regions are shown in Fig. 1. The wires have a characteristic length of 3000 Å and a periodicity of ~ 150 –200 Å in the $\langle 110 \rangle$ direction, as evidenced in the plan-view transmission electron microscopy (TEM) photograph [see Fig. 1(b)]. An encouraging feature of these spontaneously arranged SILO

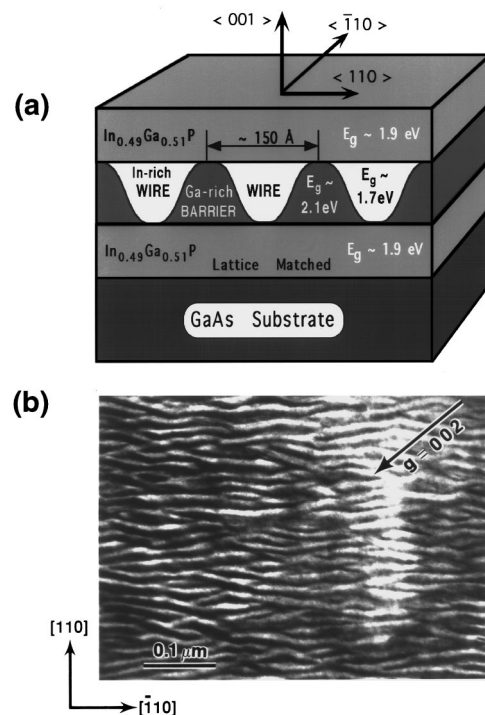


FIG. 1. (a) Idealized sample structure, with approximate band gaps of different regions as shown. (b) Plan-view TEM picture of the active region is shown. Wires have a characteristic length of 3000 Å and a minimal deviation from the preferred $\langle -110 \rangle$ direction.

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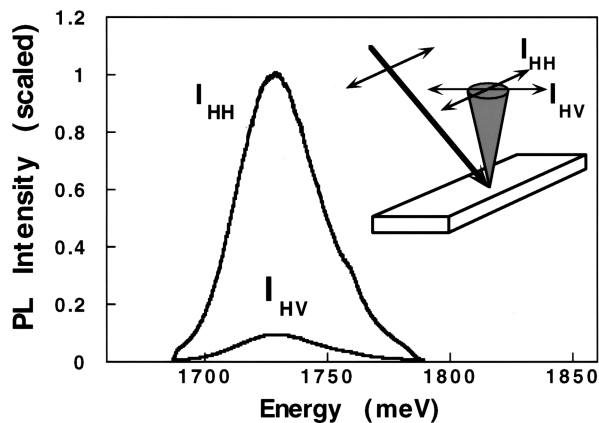


FIG. 2. Luminescence polarization anisotropy associated with the PL peak at 1728 meV. The inset shows the polarization geometries employed. The PL intensity is scaled to the maximum of the I_{HH} peak.

wires is that they are oriented preferentially in the $\langle -110 \rangle$ direction with minimal deviations.

Fourier-transform photoluminescence measurements of these samples were performed using a BOMEM DA-8 interferometer. The samples were measured at low temperature using a He flow cryostat. A polarizer and an analyzer were employed immediately before and after the cryostat, respectively. A polarization rotator was also incorporated after the analyzer. The time-resolved luminescence decay measurements were made with a photon counting setup using a Nd-YAG pumped Pyridine I dye laser with a final pulse width of ~ 4.5 ps. A SPEX triplemate spectrometer was used to disperse the PL before detection by a water-cooled micro-channel plate photomultiplier tube. The time resolution of this system is 70 ps.

Our low temperature photoluminescence measurements on the SILO grown system reveal a PL peak at 1.728 eV (Fig. 2). The average composition of the active region has an $x = 0.525$ Ga fraction. A random $\text{Ga}_x\text{In}_{1-x}\text{P}$ alloy with this composition is expected to have a band gap of ~ 1.92 eV at 4 K.⁷ The redshift of 200 meV is likely due to the large spontaneous composition modulation that leads to formation of quantum wires in this system.

Figure 2 shows our results on the polarization dependence of the QWR PL. The optical emission from these wires is highly anisotropic, with luminescence polarized along the length of the wires having an order of magnitude larger intensity than the luminescence polarized perpendicular to the length of the wires. The same degree of polarization anisotropy ($>80\%$) is also observed in absorption by the wires. We note that care was taken in our anisotropy measurements to eliminate extraneous factors that could artificially induce a polarization anisotropy. For example, we excited the luminescence with an excitation energy of 1.82 eV to ensure that the carriers are optically generated *only* in the quantum wires and in no other part of the sample structure (refer to the bandgaps in Fig. 1). Furthermore, a polarization rotator was employed so that the luminescence couples to the detection system with the same polarization for both emission polarizations.

While an anisotropic optical matrix element is often given as a sufficient condition for two-dimensional confine-

ment in a quantum wire, other effects such as strain or Cu-Pt-type ordering can also give rise to anisotropic PL and must be carefully considered. The anisotropy cannot arise solely from strain effects since the lowest bandgap regions in the Ga-rich quantum wires simply do not have enough strain to account for the large anisotropy we observe. Regarding Cu-Pt-type ordering, we have performed photoluminescence excitation spectroscopy (PLE) and PL power-dependence measurements on quantum well samples grown under similar growth conditions as SILO quantum wires. The small (<15 meV) Stokes shift of the PL peak from the PLE spectrum and the absence of an energy shift of the PL peak over 3 decades of incident power indicate that there is not a significant Cu-Pt-type ordering in these wire arrays.⁷ Finally, our contention that the large PL anisotropy we observe results from strong one-dimensional confinement is supported by energy dispersive x-ray spectroscopy⁶ and Raman spectroscopy⁸ measurements that indicate a strong confinement potential of >300 meV in the growth plane and ~ 200 meV perpendicular to the growth plane for carriers in the SILO quantum wires.

We do not observe sharp hh or lh transitions in these SILO grown wires. This is expected due to the lateral wire-width fluctuations and the confinement potential fluctuations (via the fluctuations in the amplitude of the composition modulation) in this spontaneously organized system. For this reason, luminescence decay measurements at certain discrete energies do not provide any particular insights. As an alternative, we have studied the decay of the entire PL band to qualitatively understand the carrier dynamics in SILO quantum wires.

Shown in Fig. 3(a) is the evolution of luminescence for 150 Å thick SILO quantum wires at 4 K. The PL band is truncated on the high energy side by the sharp filter cut off to block the scattered laser. Here, we have plotted the luminescence at selected delays after the excitation by a 5 ps laser pulse with 1.82 eV of excitation energy. We observe that the luminescence peaks at 1756 meV after a time of 200 ps. Since this is more than a LO phonon energy below the excitation energy, we believe that this spectrum reflects rapid carrier relaxation and exciton formation, the latter being favored in part by the large exciton binding energy in quantum wires. Within 600 ps, the PL peak shifts to 1740 meV. Deveaud *et al.*⁹ have shown that the exciton transfer times to regions with lower exciton energies are ~ 250 ps. In a more recent study, Kohl *et al.*¹⁰ have shown that the continuum excitons can relax into the energetically lowest island regions very effectively within a time that is small compared to the radiative decay times. Although these studies were performed on GaAs/AlGaAs quantum well systems, the additional dimension of confinement in our wire arrays is only expected to further reduce the transfer times. For this reason, it is likely that the 1740 meV peak that develops 600 ps after the excitation is due to the recombination of excitons that have become bound at the lowest energy sites *within* each wire. This time scale is not large enough for any significant interwire relaxation to occur.

However, looking at the PL at later delays, we observe that the luminescence peak slowly drifts to still lower energies even after 2 ns. This is an unusually long time scale for

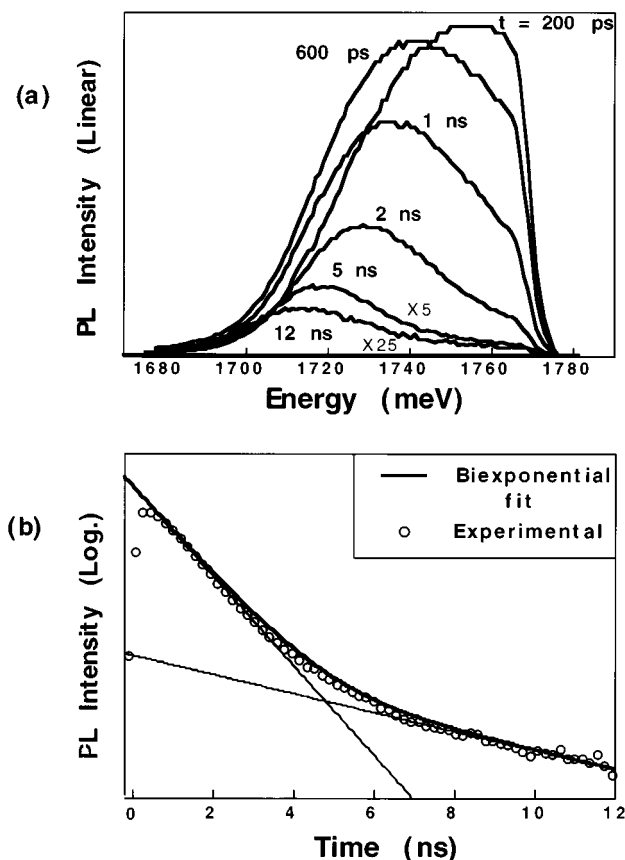


FIG. 3. (a) Evolution of the PL band after 4 ps pulse excitation. Truncation on the high energy side is due to the filter cutoff. (b) Temporal decay of the PL at 1740 meV is shown. Thin solid lines are single exponential fits to the initial and later parts of the decay and the thick solid line is the sum of the two exponentials.

any intrawire relaxation mechanism. We believe the mechanism responsible for this slow relaxation rate is the thermalization of excitons to lower energy sites in *neighboring* wires via activated hopping. The luminescence decay at 1740 meV, shown in Fig. 3(b), provides additional evidence for interwire scattering, showing an initial free exciton decay with a lifetime of 930 ps followed by a much slower bound exciton recombination with a decay time of about 4.4 ns. This behavior suggests that, due to the very high linear density ($>10^5 \text{ cm}^{-1}$) of wires in these array systems, interwire scattering plays an important role, and must be accounted for in carrier kinetics models of these systems. Notably, we have excited the sample with 1.82 eV to avoid any photogeneration of carriers in the barrier layers and, hence, have elimi-

nated any carrier capture effects. Regarding Fig. 3(b), although one would not expect to see both free and bound excitons at the same energy in a wire due to the mobility edge,¹¹ we point out that we are looking at an ensemble of wires with mobility edges at different energies. It is for this reason that we get free- and bound-exciton recombination at the same energy.

In conclusion, we observe an order of magnitude optical anisotropy in SILO quantum wires due to confinement. The large inhomogeneously broadened linewidth excludes the use of these wires from certain device applications but their high linear density (50% of active volume is wires) makes them highly desirable for other applications. Moreover, since there is no free standing surface or postgrowth processing involved, these wires are largely defect-free, as confirmed by exciton dominated long radiative decay times. We expect the active region in this system to be free from dislocations also because the strain is relaxed via composition modulation. We find evidence for slow interwire scattering at low temperatures, possibly via thermally activated hopping into neighboring wires. Long lifetimes of bound excitons could be due to reduced spatial overlap of electrons and holes at the interface fluctuations. We hope that with a better understanding of the system and the growth process, it should be possible to gain tighter control of the dimensions.

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